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Sewage sludge-to-energy approaches based on anaerobic digestion and pyrolysis: Brief overview and energy efficiency assessment

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ABSTRACT

Energy recovery from sewage sludge offers an opportunity for sustainable management of sewage sludge and energy. Anaerobic digestion and pyrolysis are among the most promising processes applicable for sewage sludge-to-energy conversion. Anaerobic digestion of sewage sludge forms methane-rich biogas, which can be utilized as fuel to offset heat and electricity consumption of the wastewater treatment sector. However, the digestion process has the limitation that it cannot sufficiently extract the energy in sewage sludge. The digested sludge is still energy profitable in that it contains considerable organic matter, but poor in biodegradability. Sludge pyrolysis is an innovative process that can convert both raw and digested sludge into useful bioenergy in the form of oil and gas, forming biochar as a byproduct that is environmentally resistant and holds potential for carbon sequestration and soil conditioning. It is expectable that sludge pyrolysis would step into practical deployment in the near future.

This paper presents a brief overview of anaerobic digestion and pyrolysis in the application to bioenergy production from sewage sludge. An assessment of energy conversion efficiency of two parallel sludge-to-energy pathways is also presented. One pathway relies on an exclusive pyrolysis process (fed with raw sludge) while the other is based on anaerobic digestion followed by pyrolysis (fed with the digested sludge). The pathway via the combination of anaerobic digestion and pyrolysis could achieve higher energy efficiency compared to the pathway employing the pyrolysis alone.

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1. Introduction

The world is facing an unprecedentedly enormous energy challenge. The rising demand for primary energy on the one hand, and the shrinking reserve of fossil fuels on the other hand, raises the global concern of how to sustain the supply of reliable and affordable energy for our future generations and ourselves. Two strategic approaches to addressing this issue are currently underlined, including exploitation and deployment of renewable energy (e.g. biomass, solar, wind, hydro and geothermal), as well as enhancement of energy efficiency in energy production and utilization. Biomass could be regarded as the most popular renewable energy resource, given that it is available locally and abundantly, technically flexible in energy production, and most attractively, it is the only renewable energy resource that can be used to produce liquid fuel.

Biomass is the term generally used to describe renewable organic-rich material that stems from various plants. Sewage sludge, which is the residue produced from wastewater treatment process sector, is rich in organic matter with composition (cellulose, hemicellulose and lignin) comparable to plant tissues. Therefore, sewage sludge is often considered as biomass [1,2]. In the EU as a whole, per capita production of sewage sludge is estimated to be 90 g per person per day [3], meaning that current annual production of sewage sludge exceeds 10 million tonnes. In China, the amount of sewage sludge produced in 2010 is estimated to reach 8.0 million tonnes [4]. In the global context, it is believed that the sludge output in the coming decades would remain to be gradually increased, considering the development of population, urbanization and industrialization.

Recently, interest in energy recovery from sewage sludge is increasing, probably because this concept positively and simultaneously addresses energy issue and the environmental concerns associated with conventional sludge treatment such as landfill and land application; sludge landfill generates undesirable emissions (e.g. leachate and landfill gas) to water, air and soil, while application of sludge to land incurs local soil contamination by heavy metals and pathogens [5].

Different technologies have been used to extract useful energy from sewage sludge, including anaerobic digestion and pyrolysis. Anaerobic digestion (AD) is thought of as one of the most technically-mature and cost-effective processes to convert sludge to methane-rich bioenergy (biogas). Great progress has been made in the understanding of the fundamental biological mechanisms and technical aspects. Historically, the AD process applied to sludge treatment is critically considered for sludge stabilization to reduce odors and pathogens. Currently, emphasis is captured on exploiting and utilizing its actual and potential ability for energy conservation and recovery. However, the risks associated with hazardous substances contained in sludge (e.g. heavy metals and POPs) cannot be alleviated via anaerobic digestion process, and the digested sludge would place impacts on the environment and on public health if necessary treatment is not implemented. On the other hand, the digested sludge is still energy profitable considering that it contains comparable organic matter.

Sludge pyrolysis is an innovative process developed to manage sludge and energy. Both bench- and pilot-scale studies have observed that approximately half the organic matter in sludge can be converted via pyrolysis into useful bioenergy (oil or gas), and that the rest of the organic matter can be dominantly distributed into pyrolytic residue (termed biochar) in a stabilized form [6,7]. It has been also observed that hazardous substances (e.g. heavy metals) retained in biochar, either transported from its precursor or captured from aqueous or soil environment, are highly immobilized [8–11]. Furthermore, land application of sludge biochar can improve soil quality and increase nutrient bioavailability [12]. These versatile advantages indicate that sludge pyrolysis will step into commercial application in the near future. Nevertheless, pursuing a high energy conversion efficiency is the key consideration for sludge pyrolysis.

It has been widely observed that both raw and digested sludge can be beneficially converted via pyrolysis into bioenergy [6,13]. This indicates that either of the following two parallel pathways is applicable for sewage sludge-to-energy conversion: one pathway is based on pyrolysis process using raw sludge as feedstock, while the other is based on anaerobic digestion (fed with raw sludge) followed by pyrolysis (fed with the digested sludge). In this study, an effort is made to evaluate and compare the energy conversion efficiency of the two pathways (Section 4). A brief overview of anaerobic digestion (Section 2) and pyrolysis (Section 3) is also presented in the case of their applications for energy conversion of sewage sludge.

2. Anaerobic digestion-based bioenergy production

Anaerobic digestion (AD) is the anaerobe-based biological process that is capable of converting biodegradable substances in the absence of molecular oxygen to biogas. Wider application of sludge digestion has being achieved, mostly motivated by its energy-related benefits; the principal benefit is that the biogas formed has a high calorific value and can be used to produce heat and electricity. Recently, an increasing attention is given to enhancing biogas production and quality (e.g. high ratio of methane to carbon dioxide). Efforts made towards this purpose primarily involve, but not limited to:

- optimization of process conditions (e.g. sludge retention time and sludge loading rate) [14,15];
- application of multi-stage process (e.g. temperature-phased and microorganism community-phased) [16,17];
- sludge pre-treatment to increase biodegradability, including chemical (e.g. acid [18], alkali [19], Fenton [20] and ozonation [21]), thermal (e.g. heating [22], freezing/thawing [23] and hydrothermal [24]), biological (e.g. enzymatic hydrolysis [25]), ultrasound [26], microwave irradiation [27] and pulse power [28].

It is important to note that, the application of the above-mentioned measures, even though being able to gain an increased output of biogas, generally requires additional energy input (heat and electricity requirement). For example, increasing process temperature can achieve more biogas yield, but meanwhile leads to more energy consumed for sludge heating. Therefore, more attention should be paid to its energy efficiency and sustainability, which requires taking into account simultaneously the two-side effects, when an energy-consuming effort to increase bioenergy yield is implemented.

Operating temperature plays a vital role in determining quantity and quality of the biogas produced, as well as the digestion rate. Three different temperature ranges, known as psychrophilic (ambient temperature), mesophilic (30–38 °C) and thermophilic (50–57 °C) range are individually adopted in the design of AD process [29–31]. The mesophilic process consistently remains dominant in practical application, principally because of its harmoniously-combined benefits with acceptable energy consumption, reliable process operation and favorable process performances (e.g. sludge reduction and biogas generation) [31,32]. The energetic performances of the mesophilic sludge digestion are summarized in Table 1.

Another important factor influencing biogasification performance of AD process is sludge retention time (SRT). The SRT is the average time that sludge is retained in digester, which can be

Table 1
Energetic performance parameters of mesophilic digestion of sewage sludge (primary data taken from [33]).

	VS destruction (% VS-input)	Specific biogas production (m^3 biogas/kg VS-destroyed)	Calorific value of biogas (MJ/m^3)	Energy yield (MJ/kg VS-input)
Range of value	40–50	0.8–1.2	15.9–27.8	
Typical value	45	1.0	25.8	11.6

adjusted by changing sludge volumetric loading rate for a volume-specific digester. It is universally observed that an increase in SRT can increase the extent of digestion reaction (generally parameterized by volatile solids reduction), leading to an increased biogas production [14,34]. For mesophilic AD system, in general SRT values below 10 days are not able to provide sufficient time for the bacteria to grow and to replace the bacteria lost in the effluent [34]. On the other hand, as indicated in previous studies [34–35], increasing SRT over 20 days can not achieve impressive improvement in volatile solids reduction and biogas production. In addition, using a higher SRT requires higher capital cost as a result of the requirement for a larger size of digester, and consumes more energy for process operation such as sludge heating and mixing. Therefore, recommended SRT values for mesophilic digestion are in the range of 10-20 days. It should be mentioned that AD process cannot completely or sufficiently mineralize organic matter in sludge. At the same time, new biomass (anaerobic bacteria) is formed during AD process. As a consequence, a substantial amount of residue known as digestate or anaerobically-digested sludge (ADS) is produced. The ADS still has the potential for energy recovery, in that it contains substantial amounts of organic matter. However, different from its original sludge, the organic matter in the digested sludge is resistant to be biodegraded. One technical option to extract energy from the bio-resistant digested sludge is to use thermo-chemical technologies such as pyrolysis, gasification and incineration.

3. Pyrolysis-based bioenergy production

Pyrolysis is the endothermic thermo-chemical process that can extract energy from organics, regardless whether the organic matter is biodegradable or not. This process has being commercialized for bioenergy production from energy crop and biomass waste [36]. All of the process products including liquid (termed bio-oil), non-condensable gaseous (termed py-gas) and solid (biochar) fractions have the potential for heat and electricity generation. Alternatively, the bio-oil product can serve as crude oil for production of transport fuels [37], while the biochar can be used as adsorbent or adsorbent precursor [38], soil conditioner [12], or used for carbon sequestration [39]. The versatile usefulness of pyrolysis products makes pyrolysis process more sustainable and beneficial compared to gasification and incineration.

Pyrolysis processes developed for energy recovery from sewage sludge can be categorized as slow pyrolysis and fast pyrolysis. Slow pyrolysis, which operates with a slow heating rate and with a long residence time, is commonly used to produce biochar or activated carbon rather than energy product (bio-oil or py-gas). Different from slow pyrolysis, fast pyrolysis undergoes a thermo-chemical process at a rapid heating rate (around 100 °C/min), which allows bio-oil or py-gas to be dominantly produced.

In terms of fast pyrolysis, there are two optional processes to produce useful bioenergy from sewage sludge. One is the moderate-temperature pyrolysis (400–550 °C) that targets bio-oil production. Currently, this pyrolysis alternative has been receiving special interest, primarily due to the facts that (1) it uses a lower pyrolysis temperature, thus requiring lower energy input for process operation, as compared to high-temperature pyrolysis; and (2) the oil produced can be easily stored and transported. Table 2 lists its energy-related performances, which were obtained by summarizing the relevant literatures that are available for calculation of energy efficiency. As shown in Table 2, about half the volatile matter in sludge can be converted into bio-oil that has a high energy content higher than 30 MJ/kg, regardless whether the sludge feedstock is subjected to anaerobic digestion or not. In some cases, the oil product possesses calorific value comparable to diesel fuels.

The other fast pyrolysis alternative is the high-temperature pyrolysis process (around 1000 °C) that is aimed to produce gaseous bioenergy (py-gas) as much as possible. We summarize the energetic performances of this alternative using available data (see Table 3). The biochar produced has low calorific value, thereby is little beneficial as energy fuel. Interestingly, this pyrolysis process can produce a higher yield of py-gas and higher hydrogen content for conversion of wet sludge than for conversion of dry sludge [43]. Increased hydrogen content is also achievable for the py-gas, when using sludge with high moisture content as pyrolysis feedstock [7]. Possible reason for these findings is, as described in previous studies [7,44], that high-temperature pyrolysis of wet sludge with high moisture content can form a steam-rich atmosphere, leading to an in situ steam reforming of the volatile compounds generated (intermediate products) and a partial steam gasification of the carbonized residue. There are a number of factors that is likely to affect the production of hydrogen-rich py-gas and its composition profile, such as sludge properties and

Table 2
Energetic performances of moderate-temperature fast pyrolysis (400–550 °C).

Sludge feedstock			Pyrolysis conditions				Bio-oil product		AEE[b] (%)	Ref.
Type[a]	VS[a] (%)	CV[a] (MJ/kg)	Heating source	Operating mode	Temperature (°C)	Time (min)	Yield (%)	CV[a] (MJ/kg)		
PS	84	23	Electric	Batch	500	20	42	37	67.6	[6]
WAS	69	19	Electric	Batch	500	20	31	37	60.1	[6]
ADS	59	17	Electric	Batch	500	20	26	37	56.6	[6]
ADS	47.0	12.3	Electric	Continuous	550		36.0	32.1	94.0	[37]
ADS	46.6	11.9	Electric	Continuous	550		27.9	31.2	73.0	[37]
ADS	38.3	8.9	Electric	Continuous	550		24.3	30.6	83.5	[37]
SS	75.5		Microwave	Batch	490	10	40	35		[40]
SS	75.5		Electric	Batch	500	30	37	30		[40]

[a] VS = volatile solids content, CV = calorific value, PS = primary sludge, WAS = waste activated sludge, ADS = anaerobically-digested sludge, SS = sewage sludge.
[b] AEE = apparent energy efficiency, calculated according to the ratio of the energy content of bio-oil product to the energy content of its feedstock.

Table 3
Energetic performances of high-temperature fast pyrolysis (around 1000 °C).

Sludge feedstock			Pyrolysis conditions			Process products		AEE[b] (%)	Ref.
Type[a]	VS[a](%)	CV[a] (MJ/kg)	Heating source	Temperature(°C)	Heating rate (°C/min)	Yield (%)	CV[a] (MJ/kg)		
SS	75.5		Microwave	1130	113	63.2 (py-gas) 7.3 (bio-oil) 29.5 (biochar)		79.3[c]	[40,41]
ADS	62.3	16.7	Electric	1000	122	54.6 (py-gas) 8.5 (bio-oil) 37.9 (biochar)	18.1 (py-gas) 36.3 (bio-oil)	59.3	[7,42]
ANDS	54.7	14.0	Electric	1000	122	51.4 (py-gas) 3.9 (bio-oil) 44.7 (biochar)	20.3 (py-gas) 36.3 (bio-oil)	74.6	[7,42]

[a] VS = volatile solids content, CV = calorific value, SS = sewage sludge, ADS = anaerobically-digested sludge, AEDS = aerobically digested sludge.
[b] AEE = apparent energy efficiency, calculated according to the ratio of the energy content of pyrolytic gas product to the energy content of its feedstock.
[c] The calculation including the energy of bio-oil.

pyrolysis conditions. Further investigation on the related reaction mechanism and end use of the py-gas remains necessary.

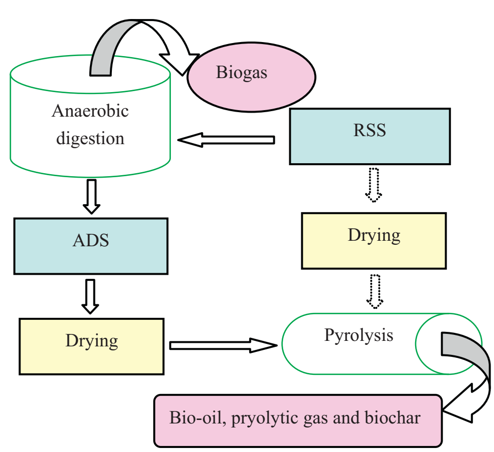
One unanticipated finding is that the energy-related performance of the high-temperature pyrolysis process is negatively dependent on the VS content of feedstock (Table 3). Although the ADS feedstock possesses a lower VS content than the AEDS, and the yield of gas produced from the ADS pyrolysis is also less than that from the AEDS pyrolysis, the pyrolysis of the ADS achieves apparent energy efficiency higher than the pyrolysis of the AEDS (see Table 3). Such observation is distinctly different from the results observed from the moderate-temperature pyrolysis, in which both the energy efficiency and the bio-oil energy output increased with the VS content of feedstock (see Table 2). This abnormity may be due to the claimed fact that, the gas from the ADS pyrolysis contained higher concentration of H_2 than that from the pyrolysis of the AEDS [7].

It should be mentioned that a pyrolysis process achieving a higher energy output (or higher apparent energy efficiency) for a given feedstock does not necessarily indicate the process is more energy-effective and affordable. The fact that pyrolysis is an endothermic thermo-chemical process implies that energy content of pyrolysis products is partly from reaction heat of pyrolysis, not just transferred from its feedstock. If the accumulative energy content of process products overwhelms the energy content of the feedstock, the energy yield of the pyrolysis might be dependent, to a great degree, on the contribution of the reaction heat. In such a case, a high energy output, which is achieved at the expense of a much higher energy input, leads to the net energy yield (or net energy efficiency) to be lowered. Therefore, more attention should be given to reaction heat of pyrolysis when considering process optimization and choice.

4. Energy efficiency assessment of two sludge-to-bioenergy pathways: a case study

4.1. Pathway description

It can be concluded from the above overviews that AD process is not able to sufficiently recover energy from sewage sludge while the digested sludge is still energy beneficial, and that pyrolysis process is applicable for the energy conversion of both undigested and digested sewage sludges. Therefore, it is possible to use either of the following parallel pathways for bioenergy production from sewage sludge (see Fig. 1). One pathway (pathway 1) is based on AD process followed by pyrolysis; raw sewage sludge is first subjected to the AD conversion with the production of methane-rich biogas and ADS (anaerobically-digested sludge), and then the ADS is processed by pyrolysis for production of bio-oil, py-gas and biochar. The other



Pathway	Bio-fuel products	Residue
Pathway 1: SS =>AD=>Pyrolysis	Biogas+ bio-oil + pyrolytic gas	Biochar
Pathway 2: SS => Pyrolysis	Bio-oil + pyrolytic gas	Biochar

Fig. 1. Flowchart of the two sewage sludge-to-energy pathways (RSS = raw sewage sludge; ADS = anaerobically-digested sludge).

(pathway 2) is based on pyrolysis conversion alone. In this alternative, the raw sewage sludge is used directly as pyrolysis feedstock.

Since raw sewage sludge before AD process contains a higher level of organic matter than the digested sludge, pathway 2 can provide a higher yield of pyrolysis products than pathway 1, but pathway 1 has an additional bioenergy product, biogas. Here, we focus the investigation on evaluating and comparing side by side the energy conversion potential of the two sludge-to-bioenergy pathways.

4.2. Methodology

The two pathways were assumed to separately convert 100 kg of raw sewage sludge (dry weight) to bioenergy. It was also assumed that the AD process introduced in pathway 1 for biogas production is a mesophilic digestion process, and that the pyrolysis process involved in the two pathways is the moderate-temperature fast pyrolysis with bio-oil as target energy product. To avoid confusion, below the term of pyrolysis refers to the moderate-temperature fast pyrolysis scenario, and the AD refers to the mesophilic digestion process.

A previous study [6] that investigated technical performances of the pyrolysis of sludges before and after AD process was revisited to identify its energetic performances (Section 4.2.1). The bioenergy production potentials from the pyrolysis of the undigested and digested sludges was thereof determined and correspondingly incorporated into the energy analysis of the two pathways. Bioenergy production potential from the AD process was deduced using mass-energy analysis based on the difference in VS value between the undigested and digested sludges (Section 4.2.2). The results obtained were merged to determine the individual energy conversion efficiency of the two pathways. The energy efficiencies are quantitatively described using two performance indicators that are described in Section 4.2.3.

4.2.1. Identification of energetic performances of the pyrolysis

To compare the energy efficiency for the two sludge-to-bioenergy pathways, basically needs to know the energetic performances (process yields and calorific value of process products) of the pyrolysis of sludges before and after AD process. In the previous study [6], three different types of sludge, named as primary sludge (PS), waste activated sludge (WAS) and anaerobically-digested sludge (ADS), were investigated under the same pyrolysis scenario for bio-oil production. These sludge feedstocks were collected from the same wastewater treatment plant. Therefore, it is reasonable to assume that the ADS is the residue of the AD process of either the PS or the WAS, or their mixture. According to the experimental findings, maximum production of bio-oil is generally achieved at 500 °C of process temperature. Table 4 lists the energetic performances at this temperature. Mass-energy balance using these data can give the pyrolysis-related bioenergy output for the two pathways.

Table 4
Characteristics of the sludge feedstock and pyrolysis products.[a]

	PS[b]	WAS[b]	ADS[b]
Sludge feedstock			
VS[b] content (wt.%)	84	69	59
CV[b] (MJ/kg)	23	19	17
Process yield of the sludge pyrolysis			
Bio-oil (%)	42	31	26
Biochar (%)	33	43	53
CV of the pyrolysis products			
Bio-oil[c] (MJ/kg-bio-oil)	37	37	37
Biochar (MJ/kg)	17	13	10

[a] The data was derived from the previous study [6].
[b] PS = primary sludge, WAS = waste activated sludge, ADS = anaerobically-digested sludge, VS = volatile solids, CV = calorific value.
[c] The calorific value of the bio-oils produced from the three sludge feedstocks was determined as approximate average value, based on the observation that the CV is independent on the type of sludge feedstock.

4.2.2. Mass and energy analysis of the AD process

The measurements of the volatile solids content of the undigested and digested sludges (PS, WAS and ADS) were used to estimate the yields of the biogas and ADS produced from the AD process of 100 kg of raw sewage sludge (PS or WAS). The estimation was conducted by mass-energy analysis with the aid of the application of empirical performance parameters of the AD process (e.g. specific biogas production and typical energy content of biogas).

The mass-energy analysis was performed using the following equations (Eqs. (1) and (2)), assuming that the dry matter of sewage sludge is a mixture of fixed solids and volatile solids, and that the amount of fixed solids in the sludge remains constant during the AD process.

$$W_{ADS} = \frac{W_{RSS} \times (1 - VS_{RSS})}{1 - VS_{ADS}}, \quad \text{or}$$

$$W_{ADS} = W_{RSS} \times (1 - R_{VS-AD} \times VS_{RSS}) \tag{1}$$

$$Y_{biogas} = SBP_{biogas} \times (W_{RSS} - W_{ADS}), \quad \text{or}$$

$$Y_{biogas} = SBP_{biogas} \times R_{VS-AD} \times VS_{RSS} \times W_{RSS} \tag{2}$$

where W_{RSS} (kg) and W_{ADS} (kg) are the dry matter amounts in the RSS and ADS, VS_{RSS} (wt.%) and VS_{ADS} (wt.%) are VS contents in the RSS and the ADS, R_{VS-AD} (%) is the reduction percentage of the VS after the AD process, Y_{biogas} (m^3) is the biogas yield, and SBP_{biogas} is the specific biogas production (SBP), which is expressed as the proportion of the biogas amount produced to the amount of the VS degraded (m^3 biogas produced/kg VS degraded).

In general, biogas production is linearly correlated with the amount of the VS degraded, when the AD process reaches stable state in operation. This indicates that values of the SBP could remain relatively stable. As previously summarized in Section 2 (see Table 1), the SPP values vary in a narrow range (0.8–1.2 m^3 biogas/kg VS degraded). This parameter can be used as a design criterion for the AD process to estimate and predict biogas yield. In this study, an average SPP value (1.0 m^3 biogas/kg VS degraded) was employed to calculate biogas yield.

The amount of Y_{biogas} obtained was used to calculate the energy output from the AD process. The calculation employed the value of 25.8 MJ/m^3 as calorific value of the biogas produced. This value was calculated at STP (standard temperature and pressure), assuming the biogas consists of 35% of CO_2 and 65% of CH_4 (the calorific value of CH_4 is 39.6 MJ/m^3).

4.2.3. Performance indicators

Two indicators including apparent and gross energy efficiencies were used to characterize the energetic performance of the two pathways. The apparent energy efficiency (*AEE*) is defined as the ratio of the energy content in the target products (bioenergy, e.g. bio-oil and biogas) to the energy content of the sludge feedstock. The gross energy efficiency (*GEE*) additionally takes into account the energy in process byproduct (e.g. biochar). The two parameters can be schematically expressed as

$$AEE = \frac{CV_{targeted-product} \times W_{targeted-product}}{CV_{sludge} \times W_{sludge}} \tag{3}$$

$$GEE = \frac{\sum (CV_{products} \times W_{products})}{CV_{sludge} \times W_{sludge}} \tag{4}$$

where $CV_{targeted\text{-}product}$ and $W_{targeted\text{-}product}$ are the calorific value (CV) and yield of targeted biofuel products, CV_{sludge} and W_{sludge} are the calorific value and dry matter amount of feedstock (raw sewage sludge), $CV_{products}$ and $W_{products}$ are the CV value and yield of all process products (biogas, bio-oil, biochar and py-gas).

Py-gas produced under the above-mentioned pyrolysis scenario mainly contains CO_2 and CO [6,37]. Previous study has found the volume proportion of CO_2 in the py-gas reaches 40–60% [37]. This indicates that the py-gas has a low energy content, and has thereby no or only limited potential for energy recovery. In this study, the py-gas product was considered energy unrecoverable and as a cause of energy loss.

4.3. Results and discussion

4.3.1. Summary of energy efficiencies of the two pathways

The results for the two pathways are summarized in Table 5. Pathway 1 has higher apparent energy efficiencies (*AEE*) than pathway 2. On average between the two RSS feedstocks (PS and WAS), overall 78% of the energy in the RSS is converted to target bioenergy (biogas and bio-oil) for pathway 1, approximately 14% greater compared to pathway 2. However, no significant difference in gross energy efficiencies (*GEE*, calculated on the basis of biogas, bio-oil and biochar) was observed between the two pathways.

The energy conversion efficiencies of the two pathways depend in part on how much bio-oil can be produced. Generally, the bio-oil production is intrinsically related to properties of sludge feedstock, and the conversion of the feedstock with a higher VS content can gain a higher yield of bio-oil. On the other hand, for a specific sludge feedstock, an increase in bio-oil yield can also be achievable through optimization of operating conditions (e.g. temperature and retention time) [45], pretreatment of feedstock (e.g. acid and base) [6], and application of catalyst [46]. Process temperature is proven to be one of the most important factors influencing product distribution and bio-oil production. Optimized temperatures for maximum production of bio-oil range from 550 °C [45] to 450–500 °C [6] and to 400–450 °C [47]. The divergence of the optimum temperature may be due to the difference in sludge origin and type, pyrolysis equipment, or even feeding mode (continuous or batch-scale).

The energy recovery efficiencies are also partly determined by energy content of the bio-oil produced. The energy contents of the bio-oils produced from three types of sludge from the same wastewater plant are found similar, with values around 37 MJ/kg, when using a fixed bed reactor in a batch-scale operating mode [6]. However, using a fluidized bed with continuous feeding, the pyrolysis of three feedstocks of anaerobic sludge from different wastewater treatment plants produced bio-oils with heating values around 31 MJ/kg [37]. These findings indicate that energy content of the bio-oil might not be dependent on properties of sludge feedstock, but is related to the pyrolysis process applied and operating conditions (e.g. equipment and operating mode).

It should be noted that the insignificant dependency between the energy content of the bio-oil formed and the feedstock itself does not indicate that other characteristics of the bio-oil are unrelated to the feedstock nature. Significant differences have also been found in viscosity and chemical composition of the bio-oil obtained from different sludge samples [37,48]. These characteristics have a substantial effect on end use of bio-oil. Thus, a comprehensive investigation of the influence of feedstock on bio-oil characteristics remains important.

4.3.2. Difference in energy efficiency between the two raw sludge feedstocks

The conversion of the two raw sludge feedstocks (PS and WAS) to bioenergy is found to be significantly different in AEE value for both pathways. In terms of pathway 2, the energy conversion only relies on pyrolysis process, while the pyrolysis conditions for the two raw sludges remain the same. The AEE difference between the two feedstocks is thereof exclusively explained by the fact that the feedstocks have different physico-chemical properties, in particular their difference in VS content. Unlike pathway 2, however, pathway 1 additionally introduces anaerobic digestion for energy conversion, aside from the pyrolysis process. Therefore, the consequence that the conversion of the PS differs from the WAS in AEE value, is related not only to feedstock itself, but also to the performance of the AD process.

In the current study, VS content in the PS (84%) is different from the WAS (69%) while their individual digested residues (ADS) have an identical VS content (59%). This indicates that the extent of VS degradation during the AD process is different between the PS and WAS. A calculation based on the mass balance (described in Section 4.2.2) can give 72.6% of VS reduction for PS and 35.3% for WAS. Such variation in the VS destruction also results in the different outcomes in energy efficiency in the case of pathway 1.

In summary, the results suggest that the energy efficiency of sludge-to-energy conversion depends not only on which conversion pathway is used, but also on characteristics of the sludge feedstock, particularly its content of volatile solids (VS). For the pathway 2 that additionally introduces the AD process, how far VS during the AD process can be reduced by the AD also plays an important role in the attempt to achieve desirable energy recovery efficiency. Below efforts were made to investigate the effects of VS content in raw sludge (Section 4.3.3) and of VS reduction level during AD (Section 4.3.4).

4.3.3. Effects of VS content in sludge

It can be observed according to Table 4 that both the energy contents of the sludge feedstocks and their bio-oil yields are linearly correlated with the VS content in sludge. The relationships can be expressed as

$$Y_{bio-oil} = 63.684 \times VS_{sludge} - 11.337 \quad (R^2 = 0.9982) \tag{5}$$

$$CV_{sludge} = 24.21 \times VS_{sludge} + 2.56 \quad (R^2 = 0.9944) \tag{6}$$

where $Y_{bio\text{-}oil}$ is the bio-oil yield (%), VS_{sludge} is the volatile solids content in sludge (%), CV_{sludge} is the calorific value of sludge (MJ/kg). Mass-energy balance calculation using these equations in combination with Eqs. (1) and (2) (see Section 4.2.2) can provide quantitative information on the influence of the VS content. Here, the calculation was carried out based on the consideration that the dry matter amount in raw sewage sludge remains constant while the relative VS content ranges from 50 to 85%, and that the VS percent reduction via the AD process is 45, 60 and 75%, respectively.

The AEE values, which were calculated without considering the energy contents of biochar and py-gas, increase with the increase of the VS content for both pathways (Fig. 2). This directly demonstrates that higher VS content of sludge feedstock is more beneficial in energy efficiency when converted into biogas and/or bio-oil. The result also indirectly reflects that, when extracting energy from sludge feedstocks with lower VS content, the accumulative energy distribution percentage of the biochar and py-gas tends to be greater. In such cases, additional attention should be paid to further exploring energy recovery potential of the biochar or py-gas.

The AEE values in pathway 1 are consistently greater than those in pathway 2, whereas differences in the AEE value between the two pathways remains unaltered, regardless of the variation in VS content of the sludge feedstock (Fig. 2). Taking the 60% of VS reduction as an example (see Fig. 2), when the VS contents of sludge feedstock ranges from 50 to 85%, differences in the AEE value between the two pathways are ca. 13.73 ± 0.34%. It should be mentioned that these results are based on the assumption that the level of

Table 5
Results of mass and energy analysis for the two pathways.

	Pathway 1: RSS[a] → AD[a] → Pyrolysis		Pathway 2: RSS → Pyrolysis
	RSS → AD	ADS[a] → Pyrolysis	
For conversion of 100 kg PS (VS[a] in the PS[a] = 84%; VS in the ADS = 59%)			
Process yields	61.0 m^3 (biogas) 39.0 kg (ADS)	10.2 kg (bio-oil) 20.7 kg (biochar)	42 kg (bio-oil) 33 kg (biochar)
Energy output (MJ)	1573.2 (biogas)	375.4 (bio-oil) 206.8 (biochar)	1554 (bio-oil) 561 (biochar)
AEE[b] (%)	84.7		67.6
GEE[b] (%)	93.7		92.0
For conversion of 100 kg WAS[a] (VS in the WAS = 69%; VS in the ADS = 59%)			
Process yields	24.4 m^3 (biogas) 75.6 kg (ADS)	19.7 kg (bio-oil) 40.1 kg (biochar)	31 kg (bio-oil) 43 kg (biochar)
Energy output (MJ)	629.3 (biogas)	727.4 (bio-oil) 400.7 (biochar)	1147 (bio-oil) 559 (biochar)
AEE[b] (%)	71.4		60.4
GEE[b] (%)	92.5		89.8

[a] RSS = raw sewage sewage, PS = primary sludge, WAS = waste activated sludge, ADS = anaerobic digested sludge.
[b] AEE = the apparent energy efficiency, GEE = the gross energy efficiency (see Section 4.2.3).

VS destruction during the AD process for different VS contents of sludge remains constant. Considering that the AD process for different VS contents of sludge could lead to different VS reduction, the effect of VS reduction should be carefully investigated.

4.3.4. Effects of VS reduction degree during AD

The effect of varying VS reduction in the AD stage on the energetic performance was also quantitatively investigated in the case of pathway 1. The quantification analysis used the same techniques as described n Section 4.3.3. The energetic performance is sensitive to the change in VS reduction in the AD stage (Fig. 2). A higher VS removal by the AD enhances the apparent energy efficiency for the pathway 1, thereby further enlarging its relative superiority over pathway 2. This enhancement, which is attributable to an increased output of biogas from the AD, on the other hand, has an impact on the biochar produced from the subsequent pyrolysis process.

To illustrate the impact of VS destruction during the AD stage on the biochar, the experimental data in Table 4 were plotted as the heating value of biochar vs. the VS content in the sludge (Fig. 3A). The variation in VS content of the digested sludge (after the AD process) is also shown, with changing VS content of the raw sludge (before the AD process), and with changing VS reduction (Fig. 3B). It can be perceived from Fig. 3B that increasing VS destruction by the AD process leads to the decrease in both yield and heating value of the biochar product. Taking the raw sludge containing 70% of VS as an example, when its VS reduction is increased from 45 to 75%, the VS contents in the resulting *ADS* would decrease from 56.2 to 36.8%; the lower VS content (36.8%) in the digested sludge leads to a lower heating value of the biochar product (Fig. 3A). Similarly, an enhancement in the VS destruction contributes to the reduction in biochar yield, as a result of the decrease in both yield and VS content of the biochar precursor (ADS).

It should be underlined that, in terms of the biochar, an earlier focus on its energy production has been turned to the current focus on its actual and potential ability in soil conditioning and carbon sequestration. When applied to soil, biochar can restore and improve soil quality, by immobilizing soil contaminants (e.g. heavy metal and POPs), retaining and gradually releasing nutrients, as

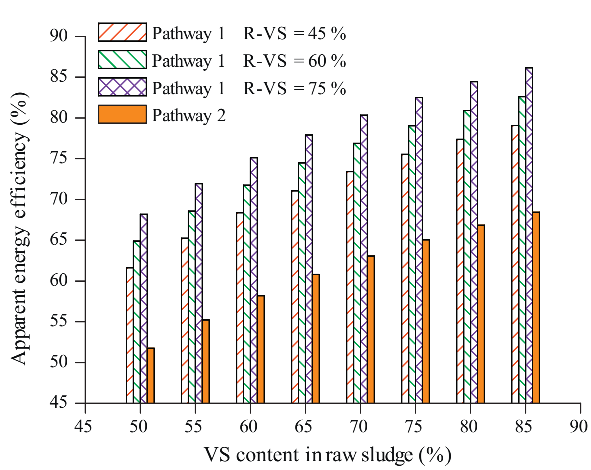


Fig. 2. Apparent energy efficiencies (AEE) as the functions of the volatile solids (VS) content in raw sludge and VS reduction level (R-VS) during anaerobic digestion.

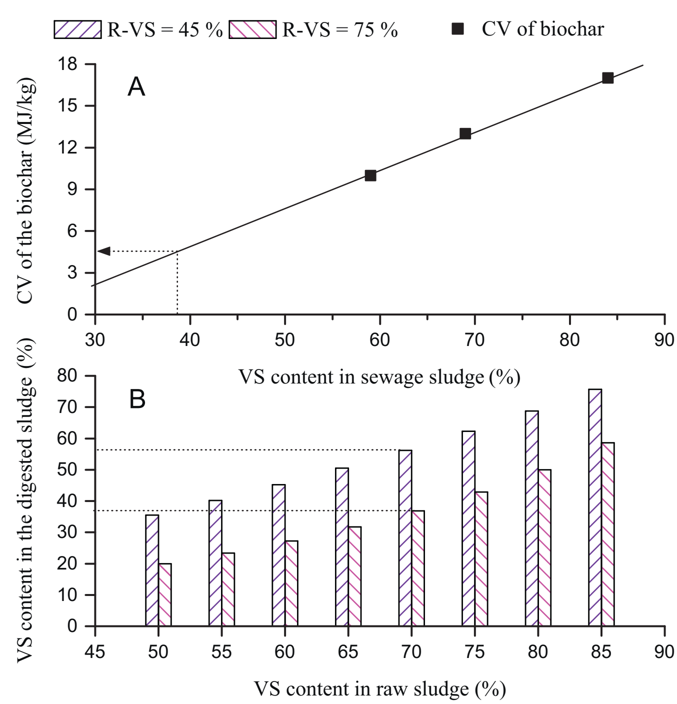


Fig. 3. (A) Calorific value (CV) of biochar as a function of the volatile solids (VS) of the sludge precursor; (B) dependency of the VS content in the digested sludge on the VS reduction (R-VS) by anaerobic digestion for the raw sludge with VS content ranging from 50 to 85%.

well as improving the soil's water-holding capacity and the habitat of microorganisms [10,49–52]. In addition, biochar is carbon rich and environmentally persistent, and its application for soil conditioning serves simultaneously as a promising approach for carbon sequestration [53].

5. Conclusions

Energy recovery from sewage sludge represents an important strategic lever for sustainable management of sewage sludge and energy. Anaerobic digestion and pyrolysis are among the most promising and sustainable processes applicable for sewage sludge-to-bioenergy conversion. Anaerobic digestion of sewage sludge, which has been well developed, can produces bioenergy in the form of biogas, which is a robust fuel that is suitable for heat and electricity generation. However, the anaerobic digestion process has the ineradicable limitation that it is not able to sufficiently recover energy from sewage sludge. The digested sludge still has considerable potential for bioenergy production on the one hand, on the other hand places a wide range of impacts on the environment and on public health if untreated or un-appropriately treated.

Sludge pyrolysis is an environmental-friendly innovative process that can successfully convert different types of sludge including primary, waste activated and digested sludges into useful bioenergy in the form of oil and gas, forming biochar as a byproduct that is environmentally resistant and can be used for carbon sequestration and soil conditioning. Practical deployment of sludge pyrolysis is expectable in the near future.

The sustainability of two alternative conversion pathways for energy recovery from sewage sludge, pathway 1 based on an exclusive pyrolysis process (fed with raw sludge) while pathway 2 based on anaerobic digestion followed by pyrolysis (fed with the digested sludge), was comparatively evaluated from the perspective of energy efficiency. It is indicated that pathway 2 can consistently achieve higher apparent energy efficiency than pathway 1. The energy recovery potential of the two pathways is strongly influenced by volatile solids content in sludge feedstock, which gradually increases with the increase of the volatile solids content. For the pathway combined anaerobic digestion with pyrolysis, a higher energy recovery efficiency can be achieved when volatile solids reduction in anaerobic digestion stage is enhanced.

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